

The resistance anomaly in the surface layer of $\text{Bi}_2\text{Sr}_2\text{Ca}\text{Cu}_2\text{O}_{8+x}$ single crystals under radio-frequency irradiation

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Abstract

We observed that radio-frequency (rf) irradiation significantly enhances the *c*-axis resistance near and below the superconducting transition of the CuO_2 layer in contact with a normal-metal electrode on the surface of $\text{Bi}_2\text{Sr}_2\text{Ca}\text{Cu}_2\text{O}_{8+x}$ single crystals. We attribute the resistance anomaly to the rf-induced charge-imbalance nonequilibrium effect in the surface CuO_2 layer. The relaxation of the charge-imbalance in this highly anisotropic system is impeded by the slow quasiparticle recombination rate, which results in the observed excessive resistance.

I. INTRODUCTION

Recently, the resistance anomaly (RA) characterized by a pronounced resistance enhancement above the normal-state value R_N near the superconducting transition has been observed in various systems such as superconducting aluminum wires^{1–3} and structures including artificial normal-metal/superconductor (NS) interfaces.⁴ Most of the previous studies on this novel phenomenon indicate that the resistance enhancement takes place either in quasi-one-dimensional structures^{1–3} or in thin films⁴ in their superconducting state when the voltage is probed within the range comparable to the characteristic length scales governing the superconducting transition. More recently, it has been found that radio frequency (rf) irradiation controls the appearance and the magnitude of the RA. This leads to explanations for the RA in terms of nonequilibrium superconductivity, such as charge-imbalance phenomenon arising either from rf-excited phase-slip centers or artificial NS interfaces, mixing of extrinsic rf noise, etc.

In this study we have observed similar resistance enhancement in *c*-axis conduction measurements on small (junction area of $25 \times 35 \mu\text{m}^2$) stacks of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ (Bi2212) single crystals irradiated by rf waves, near or below the dc superconducting transition. The RA is observed only for a three-probe measurement configuration, where the voltage measurement includes the surface CuO_2 layer which is in contact with a normal-metal (Au) electrode. The RA in this system resembles the one observed in the conventional aluminum wires² in that an excess resistance occurs near or below the superconducting transition and current-voltage (*IV*) characteristics exhibit a nonlinear excess voltage at zero bias. Since the anomaly occurs in the *c*-axis resistance of a structure including a NS interface (in this case, the interface consists of a Au electrode and the surface CuO_2 layer of Bi2212 single crystals) we believe the RA is related to the charge-imbalance nonequilibrium state produced by rf-induced “hot” quasiparticles injected from the normal-metal region into the superconducting region.¹ We assume that the rf irradiation excites quasiparticles to higher energy states above the gap of the surface superconducting layer. The enhanced accumulation of charge-imbalanced quasiparticles in the surface CuO_2 layer upon rf irradiation increases the chemical potential of quasiparticles in the layer, which can lead to the enhancement of the measured voltage across the NS interface. The charge-imbalanced nonequilibrium state is known to relax to charge-balanced state within the characteristic time $\tau_{Q^*} = (4/\pi)\tau_i[k_B T/\Delta(T)]$ at temperatures near the superconducting transition, where τ_i is the inelastic scattering time, and $\Delta(T)$ the energy gap.⁵ The resulting excess resistance due to the charge imbalance becomes most prominent near the superconducting transition, because the relaxation becomes significantly slower as the fraction of the effective states participating in the relaxation, $\Delta/k_B T$, is reduced near the transition.⁶

To our knowledge, no experimental observations of charge-imbalance phenomena of this kind have been reported before in high- T_c superconductors (HTSC). This lack of the observation of the phenomena may result from the high phonon-induced inelastic scattering rate near the superconducting transition of HTSC. The electron-phonon scattering rate $\tau_{e-ph}^{-1} (\approx \tau_i^{-1})$, depending on the quadratic power of the temperature in the two-dimensional clean limit which may be relevant to the surface conduction layer, can be a few hundred times larger in HTSC than in conventional superconductors near the transition. Thus, the probability of HTSC being in a charge-imbalance nonequilibrium state is reduced as much

and to date no RA has been reported for the materials. As we reported previously,⁷ however, the *d*-wave superconductivity of the surface CuO₂ conduction layer in Bi2212 single crystals in contact with a normal-metal electrode is suppressed due to the proximity effect. The superconducting transition temperature of the surface layer, T'_c , is reduced accordingly to at least about one-third of the transition temperature of the inner CuO₂ layers, T_c . The resultant reduced electron-phonon scattering rate near T'_c in the surface CuO₂ layer enhances the possibility of observing the nonequilibrium RA effect under rf-wave irradiation.

II. EXPERIMENT

Bi2212 single crystals were grown by the standard solid-state-reaction method.⁸ Upon cleaving a Bi2212 single crystal a 50 nm-thick layer of Au was thermally deposited on the top of the crystal to protect the surface from contamination during the further fabrication process as well as to obtain a clean interface between the normal-metal electrodes and the Bi2212 single crystal. Stacks with junction area of $25 \times 35 \mu\text{m}^2$ and the thickness of about 15 nm were then patterned using the conventional photolithography and Ar-ion-beam etching. A stack of that thickness contains about 10 CuO₂ layers. Further details of the sample fabrication are described elsewhere.⁷

The temperature dependence of the *c*-axis resistance $R_c(T)$ of a stack was measured by the conventional lock-in technique and *IV* data by a dc method. The schematic measurement configuration is shown in the inset of Fig. 1(a). All measurements were done in a three-terminal configuration with a low pass filter being connected to each measurement electrode. Three-terminal configuration was adopted to probe the surface junction by intentionally including the potential drop across the surface layer. The contact resistance between the Au electrode and the surface CuO₂ layer was less than 0.1Ω .⁹ An rf wave with frequency of 70 MHz was transmitted through coaxial cables to the current leads as shown schematically in the inset of Fig. 1(a). A capacitor was inserted between the specimen and the rf generator to separate the rf signal from the dc probing current. Because of the uncertainty in the rf coupling we were not able to determine the actual rf power transmitted to the specimen. Therefore the power levels specified below are the nominal values.

III. RESULTS AND DISCUSSION

Fig. 1(a) shows the typical *c*-axis resistance of a stack of intrinsic junctions, $R_c(T)$, in the three-probe configuration with the rf irradiation being turned on (open triangle) or off (open circle). The dc bias current was 500 nA, which corresponds to almost zero bias in the *IV* curves shown in Figs. 2(a) and 2(b). The frequency and the power of the rf wave were 70 MHz and -20 dBm, respectively. In the absence of the rf irradiation it is known that the $R_c(T)$ for a stack on the Bi2212 single crystal as illustrated in the inset of Fig. 1(a) exhibits a double superconducting transition; the upper transition at T_c ($\simeq 90$ K) is the one for the inner CuO₂ layers and the lower one at T'_c is for the top-most CuO₂ layer in contact with the Au electrode. The superconductivity in the thin CuO₂ surface layer is suppressed by the proximity contact to the thick normal-metallic Au electrode.⁷ Although all the inner layers are Josephson coupled in the temperature range of $T'_c < T < T_c$, $R_c(T)$

remains finite because the surface layer is in the normal state. The value of T'_c varies from sample to sample. It was 35 K for our specimen in this study.

When an rf wave is irradiated the $R_c(T)$ curve deviates below T'_c from the one in the absence of rf irradiation. Instead of dropping, the $R_c(T)$ keeps increasing rapidly below T'_c , before eventually showing a superconducting transition at a temperature further below T'_c . The superconducting transition for a given rf power is much sharper than in the absence of rf irradiation. The $R_c(T)$ curve above T'_c is, however, insensitive to the rf irradiation, which indicates that the excess resistance is caused by the surface CuO₂ layer. As the rf power increases the superconducting transition temperature of the surface CuO₂ layer decreases, along with an increase in the peak value of the RA as shown in Fig. 1(b). For rf power of -20 dBm the RA becomes about 7 times higher than the normal-state resistance at 100 K, which cannot be explained by local heating due to rf irradiation. Since the rf frequency used in this experiment was far lower¹⁰ than the plasma frequency ω_p the rf irradiation may have acted as a dc bias and suppressed the transition temperature.¹¹ It is also possible that the rf-induced pair breaking across the significantly reduced superconducting gap near T'_c suppressed the superconducting transition.

We also plotted (dotted line) in Fig. 1(a) the expected $R_c(T)$ at temperatures below T'_c for the normal-metal/insulator/*d*-wave superconductor (NID) junction formed by the surface CuO₂ layer in its normal state and the neighboring inner superconducting CuO₂ layer with *d*-wave symmetry.⁷ A noticeable feature is that the observed resistance enhancement near and below the superconducting transition is far above the estimated tunneling resistance for a NID-junction configuration. The RA, thus, cannot be simply explained by the equilibrium tunneling consideration.

Fig. 2(a) shows the *IV* characteristics at various temperatures below T'_c when the rf irradiation of frequency 70 MHz and power -20 dBm is on (solid line) or off (dotted line). For 29.8 K data the dotted line is not discernible because of the high overlap of the two sets of curves. Both the *IV* curves at $T = 4.2$ K and $T = 29.8$ K show DID and NID tunneling characteristics, respectively,⁷ regardless of the rf irradiation. On the other hand, near the superconducting transition (14.9 K - 23.9 K) the *IV* curves in the presence of the rf irradiation show multiple-transition features. For instance, the *IV* curve at 14.9 K shows a double transition, one below and the other above the critical current I'_c of the intrinsic surface junction in the absence of rf irradiation.

In Figure 2(b) we show a close up view of the zero-bias region of the *IV* characteristics for the three temperatures near the superconducting transition. The *IV* characteristics at $T = 18.9$ K and $T = 23.9$ K exhibit excess resistances above the normal-state resistance near the zero bias, which correspond to the RA in the $R_c(T)$ near the transition. A resistance enhancement due to the appearance of rf-induced phase-slip centers in only 0.3-nm-thick conducting layer is inconceivable. Burk *et al.* has proposed¹² that RA can be generated when the high-frequency signal is mixed with the low-frequency or dc measuring current for samples exhibiting nonlinear *IV* characteristics. Since *IV* characteristics of our sample become nonlinear near T_c of inner junctions as well as near T'_c of the surface junction, the model predicts the appearance of the RA near the two temperatures. Since the RA appeared only below T'_c in our sample, however, we exclude the possibility of external rf mixing, too.

We attribute the RA to the charge imbalance induced in the superconducting surface layer. The excess resistance R_{exc} due to the charge imbalance at an interface consisting of a

normal metal and a conventional s-wave superconductor is given by⁵

$$R_{exc} = \frac{Z(T)\tau_{Q^*}}{2e^2N(0)\Omega}, \quad (1)$$

where

$$Z(T) = 2 \int_{\Delta}^{\infty} N_s(E) \left[-\frac{\partial f}{\partial E} \right] dE, \quad (2)$$

$N(0)$ and Ω are the quasiparticle density of states of S electrode at the Fermi level in its normal state and the volume of the S electrode, respectively. $\tau_{Q^*} = (4/\pi)\tau_i[k_B T/\Delta(T)]$ is the charge-imbalance relaxation time introduced above. It should be noted, however, that Eq. (1) is applicable to a homogeneous superconducting electrode with its length much longer than the charge-imbalance relaxation length $\Lambda_{Q^*} [= \sqrt{D\tau_{Q^*}} \sim 50\mu\text{m}]$ if we assume the diffusivity D to be¹³ 31 cm²/sec].

We use Eq. (1) to estimate the excess resistance in our specimen, assuming that the charge imbalance is confined in the superconducting surface CuO₂ layer, i.e., Λ_{Q^*} is set to be the thickness of the surface layer, 0.3 nm. The assumption may be justified by the existence of a strong scattering barrier between the surface and the inner CuO₂ layers. The almost full opening of the superconducting gap in the inner CuO₂ layers in the temperature range near T'_c also reduces the probability of quasiparticle injection into the inner layers from the surface layer. For the normalized density of states of S, $N_s(E)$, we use the angle-averaged expression for the $d_{x^2-y^2}$ symmetry¹⁴ as $N_s(E) = Re \left[\frac{1}{2\pi} \int_0^{2\pi} \frac{E}{\sqrt{E^2 - [\Delta(T)\cos(2\theta)]^2}} d\theta \right]$ by assuming that the surface CuO₂ layer also has the $d_{x^2-y^2}$ -wave symmetry. Fig. 3 shows the best fit to the tail of the resistive transition as a function of reduced temperature under rf irradiation of three different power levels. In calculation we adopted $\Delta(0)/k_B T'_c = 3.85$ as obtained previously⁷ and assumed that $\Delta(T)$ follows the BCS gap equation. We also set $N(0)$ to be¹⁵ 0.8 states/[eV][Cu-atom] and adopt the nominal value for the effective volume of the superconducting electrode, $\Omega = 25\mu\text{m} \times 35\mu\text{m} \times 0.3\text{ nm}$. The inelastic scattering time τ_i is taken as the fitting parameter. The best fit values of τ_i depend on the rf irradiation power and turn out to be 2.7×10^{-8} sec, 2.0×10^{-8} sec, and 4.6×10^{-9} sec for the power of -20 dBm, -23 dBm, and -30 dBm, respectively. τ_i gets longer as the transition temperature decreases for higher rf power. Fits below the superconducting transition with only one fitting parameter τ_i turn out to be reasonably good.

The values of τ_i determined in this way appear unreasonably long compared to the ones obtained from the $R(T)$ data above T_c based on the two-dimensional superconducting fluctuation theory. For instance, the authors of Ref. [16] estimated τ_i to behave as $\tau_i = 0.75 \times 10^{-13}(100\text{ K}/T)^2$ sec for Bi2212 whiskers, which would give a few orders of magnitude shorter inelastic scattering time in the temperature range used in this experiment. As described below we believe this long inelastic scattering time stems from the two impeding mechanisms, both spatial and temporal, for effective charge-imbalance relaxation. In general, a charge-imbalanced nonequilibrium state relaxes to a charge-balanced nonequilibrium state, where pairs of conjugate electron- and hole-like quasiparticles relax to the paired ground state within the quasiparticle recombination time, τ_{rec} , which is essentially given by⁶ $\tau_r = 3.7\tau_i[k_B T/\Delta(T)]$. In addition to this temporal relaxation, in conventional

quasi-one-dimensional systems including a NS interface with homogeneous S electrode, any *local* nonequilibrium state in S can diffuse away within the characteristic length scale, Λ_{Q^*} , which is the spatial relaxation. In our highly anisotropic Bi2212 system, however, the spatial relaxation is hampered because the tunneling of quasiparticles in the surface CuO₂ layer to the neighboring inner CuO₂ layer is effectively blocked by the presence of the finite gap in the inner layer near T'_c . In addition, the recombination rate of the quasiparticles within the surface CuO₂ layer becomes significantly slower as the gap shrinks as it approaches T'_c . As a result of these two mechanisms the low-lying quasiparticle energy levels are highly occupied by the accumulating quasiparticles, blocking the further relaxation of the charge-imbalance nonequilibrium. Thus, in this situation, even the charge-balanced state is in nonequilibrium and the relaxation of the charge imbalance itself may slow down in the surface layer. It significantly enhances the charge-imbalance-induced RA in the surface layer and may explain the very long charge-imbalance relaxation time we obtained. In this sense, the quasiparticle recombination time is the bottleneck in the effective relaxation of the charge-imbalance nonequilibrium in the surface layer. Thus, one can conclude that what was actually measured in this experiment was the quasiparticle recombination time. In fact, our values from the fit above are in a reasonable range if we compare them with the quasiparticle recombination time, 0.8 ns and 2 μ s, obtained in stacks of Bi2212 single crystals by Tanabe *et al.*¹⁷ and Yurgens *et al.*¹⁸, respectively, from fits to the back-bending of *IV* characteristics, another nonequilibrium effect in dc measurements.

Although we take into account the $d_{x^2-y^2}$ -wave symmetry of the superconducting state of the surface CuO₂ layer by adopting angle-averaged quasiparticle density of states for the fit, the effect of the existence of nodes in the gap is not directly taken into account. No microscopic theory on the generation and the relaxation of the charge-imbalance nonequilibrium state for a NS interface where S has a $d_{x^2-y^2}$ order-parameter symmetry is available to date. Naive consideration may lead to a conclusion that the exsistence of nodes makes the generation of the charge imbalance easier near the node region in the k space. It may also slow down the relaxation process near the node region at any temperatures below the superconducting transition. Theoretical studies are required on the systems involving superconductors with $d_{x^2-y^2}$ -wave symmetry.

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FIGURES

FIG. 1. (a) The c -axis resistance, $R_c(T)$, in the absence (open circle) or in the presence (open triangle) of rf irradiation. The frequency and the power of the rf wave was 70 MHz and -20 dBm, respectively. The dotted line represents the equilibrium tunneling resistance of the surface junction which is assumed to be in a normal-metal/insulator/ d -wave-superconductor configuration.⁷ Inset: a schematic view of the sample geometry and measurement configuration. (b) $R_c(T)$ curves with varying rf-irradiation power, taken with the bias level of 500 nA.

FIG. 2. (a) IV characteristics at various temperatures below the superconducting transition at T'_c of the surface CuO_2 layer, in the absence (dotted line) and in the presence (dotted line) of the rf irradiation. The frequency and the power of the rf wave was 70 MHz and -20 dBm, respectively. For clarity, IV curves for different temperatures are shifted horizontally. (b) A close-up view of IV characteristics near zero bias in the presence of the rf irradiation at three different temperatures near the superconducting transition as denoted by arrows in the inset.

FIG. 3. The c -axis resistance vs reduced temperature for different rf power. The frequency of the rf wave was 70 MHz and the values of the irradiation power for the curves were -30 dBm (open circle), -23 dBm (open diamond), and -20 dBm (open triangle). The corresponding values of T'_c for the three curves were 26.3 K, 23.4 K, and 19.7 K, respectively. The solid lines are the calculation results based on Eq. (1), for the best-fit values of τ_i being 4.6×10^{-9} sec, 2.0×10^{-8} sec, and 2.7×10^{-8} sec, respectively.









